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Summary

Recent advances in the use of water and water mixtures as the dielectric in high power pulse-forming lines (PFLs) are reviewed. The important electrical properties of these liquids and their variation with mixture ratio are presented. Special attention is given to the large increase in the resistivity of water/glycol mixtures upon purification and cooling. This resistivity increase leads to a much longer intrinsic time constant than is achievable with pure water, and permits PFLs to be charged in the milli-second time frame rather than the microsecond time scales required in conventional water systems. The newly recognized phenomenon of charge injection is discussed. Finally, the impact of these findings on the design of future PFLs is presented.

Introduction

Several subsequent papers in this session will deal with specific aspects of energy storage in liquids. It is the purpose of this paper to summarize somewhat broadly the state of knowledge of the properties of water and water/glycol mixtures as they pertain to the engineering, construction, and operation of intermediate energy storage systems in the power train of pulsed power devices. We will try to atone for the attendant brevity by appending a fairly large list of references. In the past few years, considerable work has been devoted to defining the design parameters of these liquids, especially in the area of long-term energy storage. While much remains to be done, enough experience has been developed to indicate their usefulness.

We open the discussion by considering a simple prototype pulsed power device as illustrated in Figure 1.

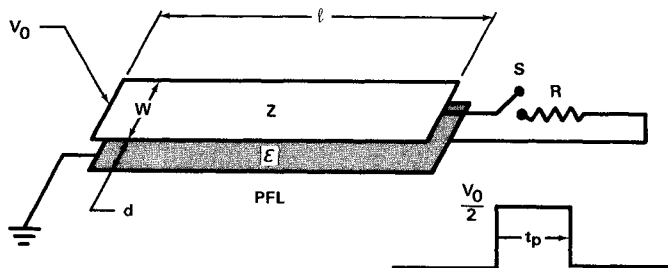


Figure 1. Prototype idealized pulsed power machine. When switch S is closed, the voltage across R is $V_0/2$ for the pulse time t_p .

The simple idealized prototype consists of a pulse-forming line, a switch, S, and a load of resistance R. The characteristic impedance, Z, of the PFL is made equal to R. The PFL is initially charged by some means

(to be discussed later) to a voltage, V_0 . For $R = Z$ (the matched condition) when the switch (assumed perfect) is closed, the voltage across the load instantaneously rises to $V_0/2$ and remains constant for a pulse time, t_p , thereafter falling instantaneously back to zero. All the electrical energy which was stored as electrical stress in the dielectric filling the PFL is delivered to the load. For simple lines of unfolded geometry, the ratio of line length, l , to pulse time, t_p , is independent of line geometry and depends only on the permittivity, ϵ , or relative dielectric constant, $\epsilon_r = \epsilon/\epsilon_0$ ($\epsilon_0 = 8.85 \times 10^{-12}$ f/m) of the material filling the PFL.

$$\frac{l}{t_p} \left(\frac{m}{ns} \right) = \frac{0.15}{\sqrt{\epsilon_r}} \quad (1)$$

Typical values of ϵ_r are: air ~ 1; insulating oils 2-6; polar liquids 20-90. In general, for pulse times from 50 to 2000 ns the use of polar liquids is indicated to avoid excessively long lines. For example, a 2000 ns simple water line is about 33 meters long; filled with oil, the line would be a few hundred meters long. For pulse time longer than 2000 ns, pulse-forming networks (PFNs) are generally preferred.

ϵ_r also appears in the equation for the characteristic impedance of the line, to be made equal to the load resistance. Z will, in general, be of form

$$Z (\Omega) = \text{GEOMETRIC FACTOR} \cdot \frac{377}{\sqrt{\epsilon_r}} \quad (2)$$

The geometric factor will depend on the shape and transverse dimensions of the PFL. For example:

$$Z (\Omega) = \frac{d}{w} \cdot \frac{377}{\sqrt{\epsilon_r}} \quad (3)$$

for the simple stripline of Figure 1. Typical high energy pulse power loads are commonly 10 ohms or less, so again high ϵ_r is desirable to maintain reasonable shape. Also note that the line will not act as a simple line unless $d \ll w \ll l$.

The scale of the transverse dimensions of the line will depend on the level of power required for a given device, e.g., for the stripline

$$P \text{ (watts)} = 6.63 \times 10^{-4} \sqrt{\epsilon_r} E_{\max}^2 \cdot wd \quad (4)$$

where E_{\max} is the maximum field which the dielectric can safely support. E_{\max} is a complicated and often not clearly defined quantity. For not only is it dependent on the dielectric, but can also be expected to depend on the electrodes (material, surface finish, area, geometry) and on the charging waveform (shape, duration). In any particular application, engineering judgment must also be used to assess the relative importance of spurious electrical breakdown. Obtaining dependable values of E_{\max} requires a large data base because of the statistical nature of the breakdown process. Improvement in this quantity by some means

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would have a large positive impact on the cost, size, weight and practicality of pulsed power devices. Optimal design of a PFL depends on a clear understanding of the design constraints. Discussion of some of these aspects may be found in References 1, 2, and 3.

With these few considerations, the common use of water in large pulsed power machines is easily understood. For not only does it have acceptable E_{\max} and high ϵ_r , it is also the least expensive and most benign of all liquids.

The above comments do not complete the picture. We have not yet considered the method of charging the line. Upon reflection, the reader will realize that the PFL serves as a power amplifying device. That is, it acts to collect and store energy over some long charging time, t_c , subsequently delivering it to the load in the pulse time, t_p . The ratio of the average power during charging to the pulsed power may be termed the power amplification (PA) of the PFL and (ideally)

$$PA = \frac{\text{PULSED POWER OUT}}{\text{AVERAGE POWER IN}} = \frac{t_c}{t_p} \quad (5)$$

For example, if the PFL collects energy over time $t_c = 10 \mu s$ and delivers that energy in time $t_p = 100 ns$, then $PA = 100$. For repetition rated pulsed power devices, obviously the maximum value of t_c is the reciprocal of the pulse repetition frequency. There are many practical advantages to maximizing the charge time of the PFL. Major advantages are lower charging source power requirements, smaller skin effect losses, and smaller transformer size requirements.

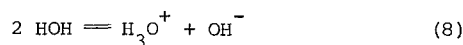
Design of the PFL charging circuit requires that another property of the liquid be considered in addition to ϵ_r and E_{\max} . This property relates to the ability of the liquid to hold charge. Thus the liquid must have a high resistivity, ρ , or equivalently, low conductivity, σ . A more convenient measure of the charge-holding capability of the liquid is the intrinsic time constant, τ given by

$$\tau = \epsilon_r \epsilon_0 / \sigma = \epsilon_r \epsilon_0 \rho \quad (6)$$

This is equal to the RC time constant of the PFL and does not depend on the geometry of the line, but only on the liquid filling it. The higher the value of τ , the less energy will be dissipated by the PFL during its charging time. It can be shown that the ratio of energy loss to ohmic heating during the charging cycle of the PFL to the energy stored is⁴:

$$\frac{\text{OHMIC LOSS}}{\text{ENERGY STORED}} = \alpha \frac{t_c}{\tau} \quad (7)$$

where α is a dimensionless constant which depends only on the wave shape of the charging waveform. Values of α range from 0.5 to 1 for common charging waveforms. τ thus gives a clear estimate of the time scale over which the charging must be completed for a particular liquid, independent of the PFL design. Unfortunately, polar (i.e., high ϵ_r) liquids tend to undergo the temperature dependent ionic dissociation reaction⁵, which maintains a level of charge carriers in even completely pure liquids. Water, for example, produces carriers from the reaction:



The carrier density from the dissociation reaction is exponentially dependent on temperature. For pure water this leads to a value of τ of 120 microseconds at $t = 20^\circ\text{C}$, rising to 670 microseconds at 0°C . Further cooling

freezes the water which destroys its dielectric constant. A value of τ in the hundreds of microseconds still does not allow efficient charging of PFL's directly from rotating machinery. Thus faced with the freezing point of water as defining an absolute maximum performance property, it becomes necessary to consider other polar liquids. The dissociation reaction, as well as severe handling difficulties, mitigate against nitrogen-containing polar liquids such as HCN. Mixtures of water and ethylene glycol, well known for their freezing point depression, immediately suggest themselves as possible candidate liquids for very high power amplification PFLs.

Water/Glycol Mixtures

Electrical Properties

Water and ethylene glycol (1,2 ethanediol) do not form simple mixtures, though they are completely miscible in each other. For example, the volume of the mixture is less than the sum of the volumes of the unmixed constituents. Upon mixing, heat is given off. The "freezing point" of the mixture is generally less than the freezing point of either substance. The term "freezing point" is placed in quotes to denote the fact that upon further cooling (as one who has ever suffered from an unexpected cold snap with insufficient anti-freeze in his automobile may have noticed), a liquid-solid slurry is formed. The mixture ratio of the solid in the slurry is not the same as the mixture ratio of the liquid, and neither is equal to the original unfrozen mixture ratio. This kind of behavior is understood by recognizing that at the molecular level the water and glycol molecules fit together in a quasi-structural arrangement which strongly depends on the interplay between the molecular dipole forces of each substance and the random thermal molecular motion.

Thus it is not to be expected that electrical properties such as the permittivity should obey a simple linear proportional relationship with mixture ratio. A half century ago, Akerlof⁶ measured the dielectric constants of water/glycol mixtures at 2 MHz for temperatures above 20°C . He discovered that the data could be accurately expressed using a simple empirical relationship:

$$\epsilon_r(T, w) = a(w) e^{-b(w)T} \quad (9)$$

where T is the celsius temperature and the empirical constants $a(w)$ and $b(w)$ depend only on the glycol weight fraction, w .

Recently⁷ we have extended these measurements to higher frequencies and lower temperatures and have found that the empirical relationship remains valid to the "freezing point" of the liquid. Additionally, to insure that the cooling did not slow the molecular dipole response time to such an extent as to compromise the utility of these mixtures for the few nanosecond risetimes characteristically required for pulsed power machines, we measured the complex dielectric constant of the mixtures. In the framework of simple Debye theory^{8,9} the complex dielectric constant is given by:

$$\epsilon_r(\omega, T, w) = \epsilon_\infty(w) + \frac{\epsilon_s(T, w) - \epsilon_\infty(w)}{1 + j\omega\tau_D(T, w)} \quad (10)$$

Here, $\epsilon_\infty(w)$ is the dielectric constant at infinite frequency, and taken equal to the square of the index of refraction. $\tau_D(T, w)$ is the Debye relaxation time and is the measure of the time required for the dipoles to

realign in a changing field. The Debye formulation was found to summarize the data very well for frequencies up to 108 MHz. The measured values of $\epsilon_s(T,w)$ and $\tau_D(T,w)$ are shown in Figures 2 and 3. Thus for frequencies to 108 MHz, $\epsilon_r(\omega,T,w)$ is substantially the same as $\epsilon_r(2\text{ MHz},T,w)$. It is also seen that while $\tau_D(T,w)$ does show exponential increase with decreasing temperature, it remains less than a nanosecond and hence still a small factor.

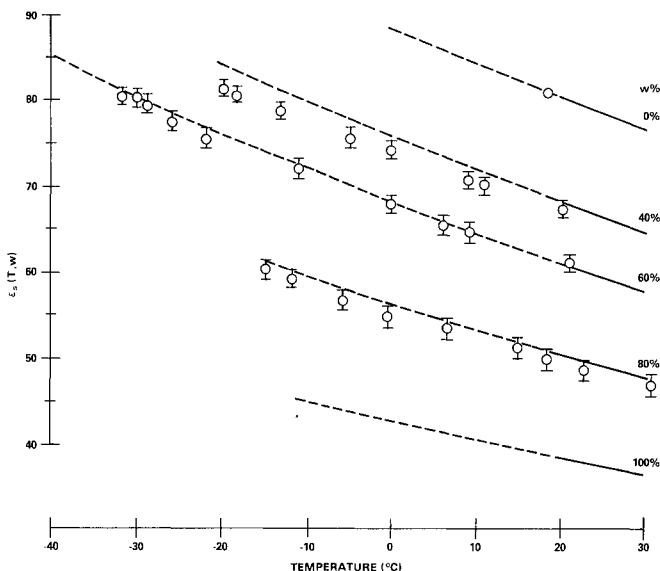


Figure 2. Static dielectric constant of glycol/water mixtures. Dashed lines are extrapolations using Akerlof's formulation, Equation (9).

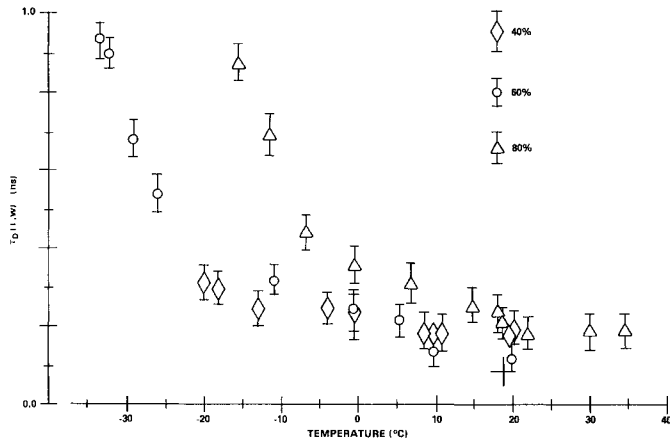


Figure 3. Debye relaxation time of glycol/water mixtures. This is a measure of the time required for the dipoles to reorient; thus, it is an ultimate minimum risetime of a PFL.

The measurements of the intrinsic time constant of purified, cooled glycol mixtures showed the anticipated dramatic increase upon cooling. Figure 4 shows the results of the measurements. A simple empirical formula of the same functional form as Equation (9) can be used to summarize the data:

$$\tau(T,w) = \tau(O,w) e^{-c(w)T} \quad (11)$$

where $\tau(O,w)$ and $c(w)$ are empirical constants analogous to $a(w)$ and $b(w)$ in Equation (9).

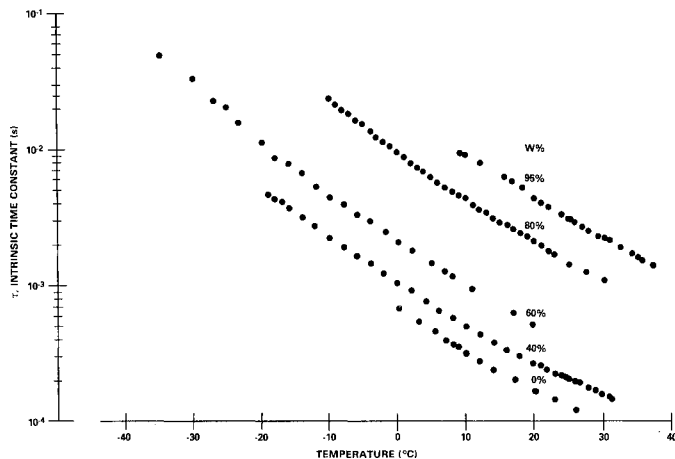


Figure 4. The intrinsic time constant of glycol/water mixtures.

Table I lists various physical properties of water/glycol mixtures with values for the empirical constants of Equations (9) and (11). This table can be used for design studies. The empirical constants for $\tau(T,w)$ are accurate to only about 10%. The actual variation with temperature is somewhat stronger than the simple exponential fit due to decrease of the ionic mobilities.¹⁰

Charge Injection

The achievement of these long intrinsic time constants for the cooled glycol mixtures has uncovered a new decay mechanism which dominates intrinsic decay at high fields. This new decay is termed charge injection as it arises from the injection and transport of net space charge into the dielectric medium. Charge injection is mathematically more complicated than simple intrinsic decay, for it not only depends on the dielectric liquid, but also upon the electrode material, electrode spacing, and the external charging circuitry. The injected charge may come from only one (unipolar charge injection) or from both (bipolar charge injection) electrodes. The charge injection dominated decay can be characterized by the transport time of the injected charge across the electrode gap. This time, t_{tr} is (for parallel flat plates separated by d):

$$t_{tr} = \frac{d}{\mu E} = \frac{d^2}{\mu V} \quad (12)$$

where V is the voltage across the plates, $E = V/d$ is the field and μ is the mobility of the injected charge.

When charge is injected, the field in the dielectric is not constant. The situation is depicted in Figure 5. The governing equation shown in the figure for the decay of the field between the plates is a partial differential equation which, except for certain special conditions, requires numerical solution. Charge injection is manifested in the term containing $\partial E/\partial x$. A more detailed mathematical theory of charge injection as it pertains to cooled glycol systems may be found in Reference 11. Charge injection, it should be realized, probably occurs in all liquid systems; it is only when $t_{tr} \ll \tau$ that it dominates the voltage decay. Figure 6 shows the difference in high voltage decay waveforms when an isolated, charged capacitor decays (1) predominantly from intrinsic decay, and (2) predominantly from charge injection decay. Electrodes in pure water charge inject, but because normally

w (%)	D_4^{20}	"f.p." C	Equation (9)		Equation (11)		$\epsilon_{\infty}(w)$ (n_D^2)
			a(w)	$\frac{b(w)}{10^{-3}/C}$	$\tau(o,w)$ ms	$\frac{c(w)}{10^{-2}/C}$	
0	0.9982	0.00	88	4.72	0.67	6.7	1.77
20	1.0241	-7.90	82	4.95	--	---	1.83
40	1.0514	-23.80	76	5.30	1.05	8.1	1.88
60	1.0765	-51.23	68	5.53	2.07	8.5	1.94
80	1.0927	-20.00 ^{est}	57	5.48	9.40	9.1	1.99
>95	1.1080	-11.50	43	5.16	19.60	8.0	2.04

Table I. Properties of water/glycol mixtures.

$t_{tr} > \tau$ for water, it is not as readily apparent as in the cooled water/glycol systems. Zahn and co-workers^{12,13,14} have made extensive study of charge injection in water using Kerr effect measurements. The Kerr effect shows directly the space charge buildup and transport in the liquid from the injecting electrodes. Recent results on charge injection studies are presented in two papers in this session.^{15,16}

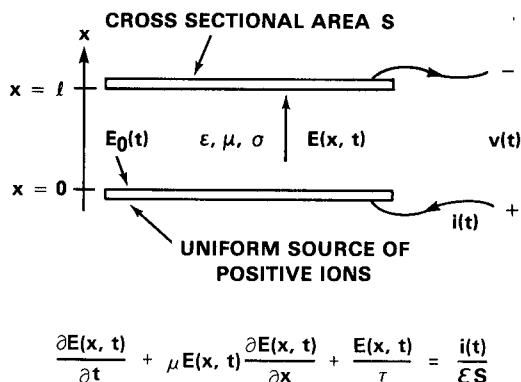


Figure 5. Charge injection modeling. Except in certain specialized circumstances, the partial differential equation requires numerical solution.

Electrical Breakdown

Based on limited experiments, we found that the electrical strength of the water/glycol mixtures is as good as, and possibly better than, cooled purified water. In Table II we list some results in terms of three figures of merit for electrical strength: E_{max} , W and A , where:

$$W = \frac{1}{2} \epsilon E^2$$

$$A = W \cdot t_{eff} \quad (13)$$

t_{eff} is defined in the same way as for short pulse testing, i.e., it is the time for which the impressed voltage exceeds 63% of the maximum impressed voltage. More convincing experiments with larger areas to produce a much larger data base are scheduled to be undertaken this year.

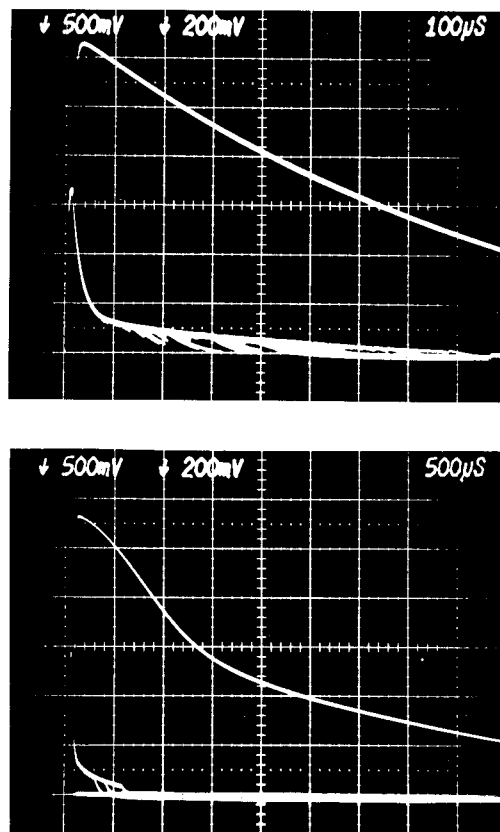


Figure 6. High voltage decay between parallel plates in an 80% glycol mixture. The plate area is 81 cm^2 with 5.1 mm spacing. The initial field is 180 kV/cm. Top oscillograph: Normal exponential decay typical of intrinsic dominated decay. Bottom: Charge injection dominated decay. Note that the waveform is no longer exponential and the difference in time scale of each picture. Top 100 $\mu\text{s}/\text{div}$, bottom 500 $\mu\text{s}/\text{div}$. Top is for $\tau < t_{tr} \sim 1 \text{ ms}$. Bottom shows decay when $\tau > t_{tr}$. The sharply peaked waveforms under each decay waveform are Marx charging signals and not of interest here.

w (%)	T (°C)	ϵ_s	τ (ms)	t_{eff} (ms)	E_{max} (MV/m)	W_{max} (kJ/m ³)	A (J s/m ³)
0	0	88	0.67	0.25	13	66	16
40	25	67	0.20	0.10	16	76	8
	-11	79	2.70	0.40	16	90	36
60	30	58	0.30	0.18	16	66	20
	-23	77	15.00	0.97	14	67	65
80	25	49	1.40	0.45	21	96	43
	-10	60	23.00	1.00	17	77	77
95	28	40	2.50	0.20*	27	129	26

*Limited by external impedance.

Table II. Electrical breakdown strength of glycol/water mixtures

Engineering Implications

Based on the time scale in which the PFL is charged, three classes of pulsed power machines may be roughly defined: (1) Very fast charged, (2) fast charged, and (3) slow charged. Typical power trains for these three classes are shown in the block diagram, Figure 7.

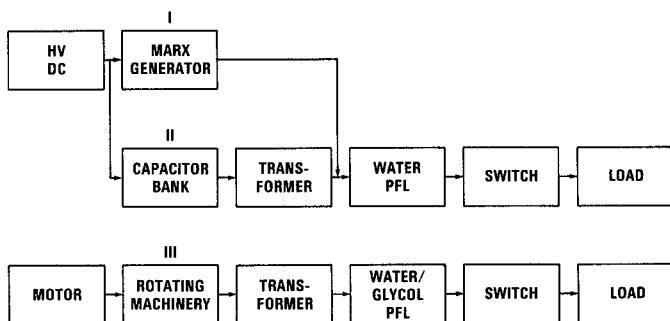


Figure 7. Power trains for three classes of pulsed power machines.

Class 1 - The very fast charged pulsed power machines are characterized by charging time scales around 1 microsecond. The PFL charging is commonly done from a Marx bank, serving as a first intermediate energy store and also yielding voltage gain. The power amplification in the PFL is modest, typically about 10. Because the charge is on the PFL for such a short time, only nominally pure, uncooled water ($\rho = 2.5 \text{ Mohm-cm}$, $\tau = 15 \text{ } \mu\text{s}$) is required. These devices, because of the Marx spark gaps, are limited to very low rep rate operation. Energy efficiency is not a factor. In this time scale, the Martin Relation¹⁷:

$$E_{max} t_{eff}^{1/3} = \frac{K}{A^{0.1}} \quad (14)$$

is operative. The effective energy density in the PFLs can be large because E_{max} is higher.

Class 2 - The fast charged machines are characterized by PFL charging times from 2-100 microseconds. The charging is commonly done by discharging a conventional capacitor bank, ringing up the voltage in the PFL via an inductor or transformer. The PA in the PFL is from 10-1000. This class of machines requires the use of very carefully purified water, and for charging

times in excess of 15 microseconds, requires very pure cooled water. In future rep-rated systems, the first intermediate storage capacitors may be replaced by cooled glycol/water systems.

Class 3 - The slow charged machine is a new class of pulsed power device charged in times exceeding a millisecond. The slow charging of the PFL can be accomplished with turbine-driven rotating machinery (alternator, compulsator) ringing up through a transformer. The power amplification in the PFL can exceed 100,000. The power train requires no first intermediate energy store. This class of machine will require the use of very pure cooled water/glycol mixtures. In terrawatt peak power, megawatt average power machines, energy efficiency is of prime concern. Ohmic losses and other inefficiencies can be expected to produce waste heating near the megawatt level. Flowing liquid heat transfer systems will be required. Glycol/water systems are excellent heat transfer agents, suggesting that it may be desirable for the entire power train to be immersed in the cooled, purified, liquid dielectric.

Liquid conditioning techniques for the water/glycol mixtures are generally the same as presently employed for high purity water systems.¹⁸ Cooling a cubic meter of glycol/water mixture from 20°C to -35°C requires removal of more than 200 MJ of heat; so for large systems, this is a time-consuming effort. The required temperatures are well within present day refrigeration technology. Our experience has been that the deionizers require less volume throughput at low temperature to maintain purity because the mixtures become less active ionic solvents. Deaeration (necessary for bubble control and achievement of highest purity) should be accomplished before cooling. This process is somewhat more difficult for the high glycol fraction mixtures due to their increased viscosity. Long holdoff time is now limited by charge injection decay phenomena, not intrinsic decay. Some advances in this field will be reported in following papers.

The energy storage cost is presently 2-3¢/Joule for the glycol alone. This is still a small part of the system cost (electrode structure, etc.). The energy densities presently achievable ($\sim 50 \text{ mJ/cm}^3$) are not spectacular. The appeal of these systems lies in their simplicity and repairability. Big payoffs in cost and size can occur if means of increasing the threshold breakdown field are developed.

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